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(6) GENERATION OF COHERENT VUV AND SOFT X-RAYS.

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I. INTRODUCTION

The goal of this program is the development of practical sources of coherent vacuum ultraviolet radiation. During this reporting period ~~our~~ The primary effort has been on the development of a new type of VUV light source based on spontaneous anti-Stokes scattering. ~~We believe~~ this type of source may prove bright enough to serve as a pump for very short wavelength lasers. Details of ~~our~~ The work are ~~presented in Section II~~ included.

In addition, ~~we have completed~~ ~~our~~ an investigation of factors limiting the efficiency of 3547 Å ~~→~~ to 1182 Å generation in Xe was completed; a manuscript describing ~~our finds~~ The results has been accepted for publication the the IEEE Journal of Quantum Electronics, and is included as Appendix A.

II. DEMONSTRATION OF ANTI-STOKES VUV LIGHT SOURCE

(L. J. Zych, J. Lukasik, J. F. Young, and S. E. Harris)

The increasing difficulty in generating soft x-ray laser radiations using parametric processes has led us to investigate a different approach to the problem. Recently we have developed a new type of VUV or soft x-ray light source with quite unique properties. We intend to use this light source to innershell ionize an alkali metal; thus, produce a population inversion necessary to create a soft x-ray laser.

The light source, proposed by Harris,[1] was constructed by rapidly transferring metastable population utilizing spontaneous anti-Stokes scattering. The source is narrow band, has a short pulsewidth, and is of very high brightness.

In recent experiments we have used a glow discharge to store population in the $2s\,^1S$ level of He at $\omega_2 = 166{,}278\ \mathrm{cm}^{-1} \cong 601$ Å. For typical experimental conditions, the density of (stored) excited state atoms is $N_2 \cong 3 \times 10^{11}$ atoms/cm^3, while the ground state density is $N_1 \cong 10^{17}$ atoms/cm^3. An incident Nd:YAG pumping laser of wavelength 1.064 μ was focused into the glow discharge. Generated VUV radiation was observed at both the upper sideband $\omega_{VUV} = \omega_2 + \omega_p = 569$ Å (the anti-Stokes sideband), and at the lower sideband $\omega_{VUV} = \omega_2 - \omega_p = 637$ Å (Fig. 1).

We note that laser induced two-photon and anti-Stokes scattering in the VUV has previously been observed by Braunlich and Lambropoulos.[2] In their experiments a collimated atomic beam of deuterium metastables was generated

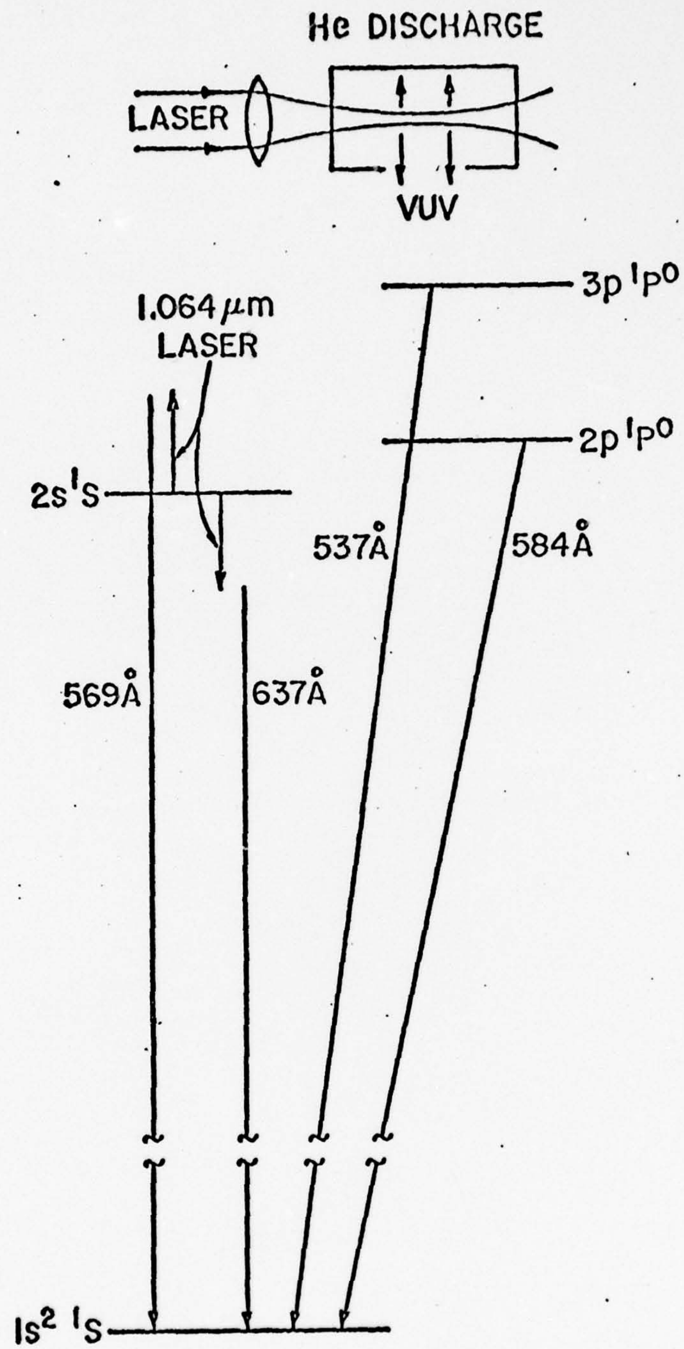

Fig. 1--Schematic and energy level diagram for the first two resonance lines of He and for the spontaneous anti-Stokes light source. An upper and lower sideband is obtained.

by charge exchange with Cs, and upper and lower sideband radiation was observed at 1090 Å and 1373 Å. Since both the metastable and the ground state densities in these beam experiments were about 10^6 atoms/cm^3, the medium was optically thin at the radiated frequency, and the emitted radiation was very weak.

The anti-Stokes with two-photon emission may be described in terms of a laser induced (atomic) emission rate $A(\omega)$, and the laser induced absorption cross section $\sigma(\omega)$. For the range of laser power densities of interest here, both $A(\omega)$ and $\sigma(\omega)$ increase linearly with power density and are related to each other in the same manner as are the emission and absorption coefficients for single-photon processes. The brightness of the light source $B(\omega)$ (radiated power per area per sterradian per bandwidth) is determined by the interplay of the emissive and absorptive processes; and for an infinitely long cylinder with outer radius r_0 is given by

$$B(\omega) = \frac{1}{4\pi^3} \frac{\hbar\omega^3}{c^2} \left[\frac{1}{\exp\,(\hbar\omega_{21}/kT) - 1} \right] \left\{ 1 - \exp\,[-\,\sigma(\omega)(N_1 - N_2)r_0] \right\} \quad (1a)$$

$$\sigma(\omega) = \frac{\pi\omega}{6c^2\epsilon_0^2\hbar^3} \left[\sum \left(\frac{\mu_{2i}\mu_{i1}}{\omega_i - \omega_{VUV}} + \frac{\mu_{2i}\mu_{i1}}{\omega_i + \omega_{VUV}} \right) \right]^2 \frac{P_p}{A}\, g(\omega - \omega_{VUV}) \quad (1b)$$

T is the temperature of the metastable level, i.e., $N_2/N_1 = \exp - (\hbar\omega_{21}/kT)$; μ_{ij} are the matrix elements; ω_i are the frequencies of the intermediate states; P_p/A is the power density of the pump laser (mks units); and $g(\omega)$ is the (normalized) lineshape for the two-photon absorption of the generated radiation ω_{VUV} .

In the optically thin case, i.e., $\sigma(\omega)(N_1 - N_2)r_0 \ll 1$, $B(\omega)$ increases linearly with the laser power density and is the same as obtained from the usual spontaneous scattering cross section point of view. As the laser power density is increased to render the media nominally two-photon opaque, i.e., $\sigma(\omega)(N_1 - N_2)r_0 = 1$, the brightness approaches that of a blackbody radiator at the temperature T of the metastable level.

The glow discharge was obtained in a 4 mm inner-diameter quartz tube with a 0.9 cm long and 200 μ wide slit cut into its sidewall. The input slit assembly of a McPherson 225, normal incidence spectrometer was removed and the glow discharge tube was mounted flush against the spectrometer so that the tube was at the exact position of the input slits. The discharge tube had a total length of 30 cm, a cylindrical cathode, and a pin anode at opposite ends. The pressures ranged from 0.7 to 6 torr and the typical discharge current was 120 ma. A pressure of less than 7×10^{-5} torr was maintained at all times inside the McPherson spectrometer.

The Q-switched Nd:YAG laser having a peak power of 1 MW, a pulse length of 8 ns, a linewidth of about 0.75 cm^{-1}, and a repetition rate of 10 pps, was focused into the center of the glow discharge tube to a spot size of $\pi\omega_0^2 \cong 10^{-4}\ cm^2$ and a confocal parameter $b \cong 2$ cm ; this produced a power density of about $10^{10}\ W/cm^2$ in a filament parallel to the slit.

Detection was accomplished with a sodium salicylate screen followed by a RCA 7265 photomultiplier, a 2 ns rise time amplifier, a pulse height discriminator, a coincidence gate, and a counter. Typically, 30 counts per minute were registered. This small count rate was the result of the 10^{-7} duty cycle of the pumping laser, and a ratio of photons detected to total VUV photons generated of 10^{-8}, i.e., under typical conditions the source, when firing, emitted photons at the rate of about 10^{15} photons/sec. Noise counts produced by the visible emission of the glow discharge limited the signal-to-noise ratio to 15. The coincidence gate, set to a width of 45 ns, was triggered by the Nd:YAG laser.

The first row of Table I shows the measured relative intensities of the laser induced upper sideband at 569 Å, the lower sideband at 637 Å, and the two He resonance lines at 537 Å and 584 Å. No other emissions were observed in the range of 500 Å to 700 Å. In the second row of Table I we estimate the spectral brightness of each of these lines. To do this, the linewidth of the laser induced emission was assumed to be 1 cm^{-1} (Doppler broadening), while that at 537 Å and 584 Å was taken as 0.1 Å and 0.5 Å respectively based on linewidth calculations. Also, a geometrical factor of 3.5 was included to account for the larger effective area of the resonance line radiation.

The application of the anti-Stokes source to the problem of soft x-ray laser construction is based both on its narrow linewidth and short time scale. To obtain a gain of e^{10} in a 1 m path length in a Duguay-Rentzepis innershell ionized laser operating on the 697 Å transition of Rb^+ requires an inversion of 4.6×10^{11} $atoms/cm^3$ and for confocal focusing a deposited energy of only 1 μJ. The narrow linewidth of the anti-Stokes source, when tuned to a high-lying state for innershell excitation, should allow a large cross section for absorption and abrupt stopping of the incident radiation. The short rise time of the anti-Stokes source should allow the process to be completed in a time short compared to the spontaneous decay time of the excited ion.

We are presently continuing our measurements of properties of this source and comparing our results to Eq. (1a) as a function of pump power density and pressure.

TABLE I

Comparison of the Laser Induced Emission to He Resonance Lines

	RESONANCE LINES		LASER INDUCED EMISSION	
	537Å	584Å	569Å	637Å
OBSERVED COUNT RATE 10^6 COUNTS/sec	0.330	13.00	6.70	0.44
ESTIMATED BRIGHTNESS $10^{15}\,\frac{\text{PHOTONS}}{\text{sec cm}^2\text{ sr cm}^{-1}}$	0.015	0.35	2.00	0.13

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2. P. Braunlich and P. Lambropoulos, Phys. Rev. Lett. 25, 986 (1970).

III. FUTURE EFFORT

We expect future effort to be primarily devoted to the development and characterization of the new anti-Stokes VUV source, especially the verification of the blackbody characteristics.

IV. PUBLICATIONS

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APPENDIX A

LIMITATION OF 3547 Å TO 1182 Å CONVERSION EFFICIENCY IN Xe *

by

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ABSTRACT

We report experiments which indicate that the 3547 Å → 1182 Å conversion efficiency in Xe-Ar mixtures is limited by Kerr-induced dispersion to about 0.9%. The mixed-frequency third order nonlinearity $\chi^{(3)}(-3\omega, 3\omega, -\omega, \omega)$ significantly alters the index of refraction at 1182 Å in the presence of large power densities at 3547 Å, affecting phasematching. We suggest using a Xe-Mg-Ar mixture to reduce the effect, thus permitting increased efficiencies.

LIMITATION OF 3547 Å TO 1182 Å CONVERSION EFFICIENCY IN Xe

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The recent demonstration of large gains in discharge-pumped XeF for single 30 ps 3547 Å pulses[1] indicates that very high power, high energy sources of 3547 Å radiation should be practical. This fact has motivated us to explore the factors which limit the ultimate efficiency of the tripling process 3547 Å → 1182 Å in Xe phasematched with Ar, as first demonstrated by Kung, Young, and Harris.[2] Clearly, high efficiencies coupled with amplified high power 3547 Å pulses could lead to a powerful source of 1182 Å radiation for microlithography, holographic microscopy,[3] and for the generation of extremely short wavelengths using additional nonlinear processes.[4]

The measurements of Ref. 2 indicate that 3547 Å → 1182 Å conversion efficiencies of several percent should be possible for reasonable experimental parameters, for example a 3 mJ, 30 ps pulse of 3547 Å focused to a 10 cm confocal parameter in a mixture of 6 torr Xe and 63 torr Ar. However, despite the work of Ref. 2, and subsequent efforts in our laboratory, observed efficiencies have reached only about 0.2%. The cause of the low efficiencies has been attributed to a number of factors, including loss, poor gas mixing, and bad laser mode quality. In this paper we report experiments which indicate that the conversion efficiency is in fact limited by Kerr-induced dispersion. In particular, the dominant effect appears to be produced by a mixed-frequency Kerr nonlinearity $\chi^{(3)}(-3\omega, 3\omega, -\omega, \omega)$ which significantly changes the index of refraction at 1182 Å in the presence of large power densities at 3547 Å, thus affecting phasematching. After presenting evidence to support this conclusion we propose a possible method of compensating the Kerr-induced dispersion and substantially increasing efficiencies.

Our experimental set-up is similar to that reported in Ref. 2. A single ~ 30 ps pulse selected from the output of a passively mode-locked Nd:YAG oscillator is amplified by a Nd:YAG amplifier chain, frequency doubled in angle-tuned KH_2PO_4, and mixed with the remaining 1.06 μm radiation in angle-tuned KH_2PO_4 to produce 3547 Å. This radiation is focused to a confocal parameter of $b = 1$ cm at the center of a 50 cm gas cell; the resulting 1182 Å light passes through the LiF output window into a calibrated acetone ionization chamber. The 3547 Å → 1182 Å conversion efficiency is measured at various values of input power density as the number density, N cm^{-3}, of the gas is slowly increased. Both pure Xe and known mixtures of Xe and Ar were used; mixtures having a desired Xe:Ar ratio of 1:R are prepared at least 24 hr. before a measurement and allowed to equilibrate.

In the presence of the Kerr effect the k vector mixmatch, $\Delta k = k(3\omega) - 3k(\omega)$, of the mixed gas can be expressed as

$$\Delta k = \frac{6\pi}{\lambda}\left(\alpha + \beta R + \frac{\gamma P}{A}\right)\frac{N}{1 + R} \qquad (1)$$

where λ is the fundamental wavelength in cm, P/A is the power density in W/cm^2 at 3547 Å, and α and β are the index mismatch per atom of Xe and Ar respectively. For this process Xe is negatively dispersive $(\alpha < 0)$, while Ar is normally dispersive $(\beta > 0)$. We have assumed that only Xe atoms contribute to the Kerr effect and have defined a Kerr-induced index mismatch per atom as

$$\gamma = 0.079\ [\chi^{(3)}(-3\omega, 3\omega, -\omega, \omega) - \frac{1}{2}\chi^{(3)}(-\omega, \omega, -\omega, \omega)] \quad \text{(esu)} \qquad (2)$$

For tight focusing the conversion efficiency can be expressed as[5]

$$\mathcal{E} = (1.5 \times 10^{-5})\ [\chi^{(3)}(-3\omega, \omega, \omega, \omega)]^2 \frac{(P/A)^2}{(\alpha + \beta R + \frac{\gamma P}{A})^2} G(b\Delta k) \quad \text{(esu)} \quad (3)$$

where the focusing factor G has a peak value of 46.3 for $b\Delta k = -4$ or for

$$N_{peak} = \frac{-2\lambda}{3\pi b} \frac{1 + R}{(\alpha + \beta R + \frac{\gamma P}{A})} \qquad (4)$$

Note that such a peak exists only if the net dispersion of the medium is negative, or $R < (-\alpha - \gamma P/A)/\beta$.

Figure 1 shows measured values of the peak conversion efficiency as a function of P/A for pure Xe, while Fig. 2 shows the pressure of Xe at which each maximum occurred. For low values of P/A the optimum Xe pressure is constant at 7 torr which implies $\alpha = -3.3 \times 10^{-23}$ cm^3; similar measurements with various Xe:Ar mixtures indicate that $\beta = 2.8 \times 10^{-24}$ cm^3.

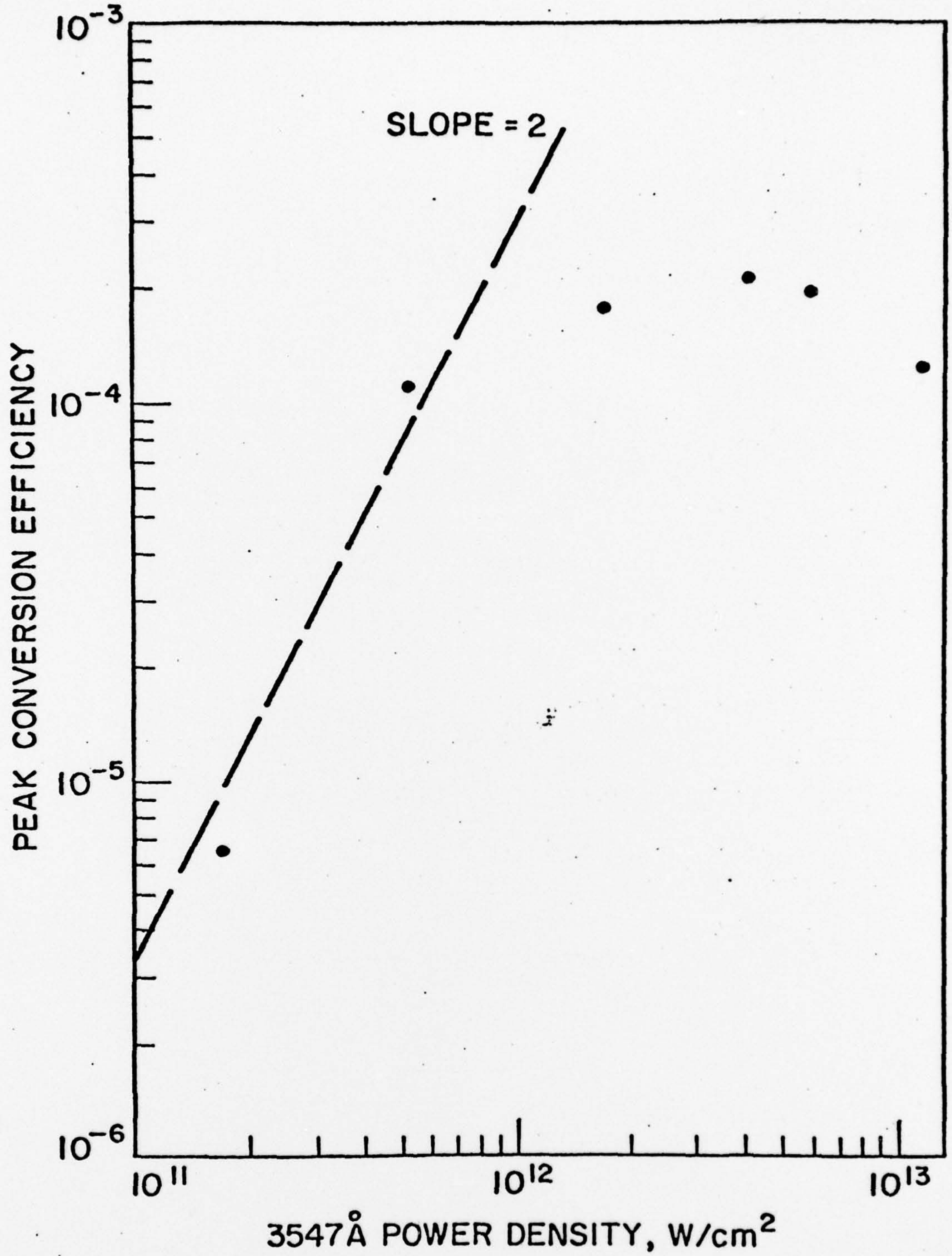

Fig. 1--Measured 3547 Å → 1182 Å peak conversion efficiency vs. fundamental power density in pure Xe.

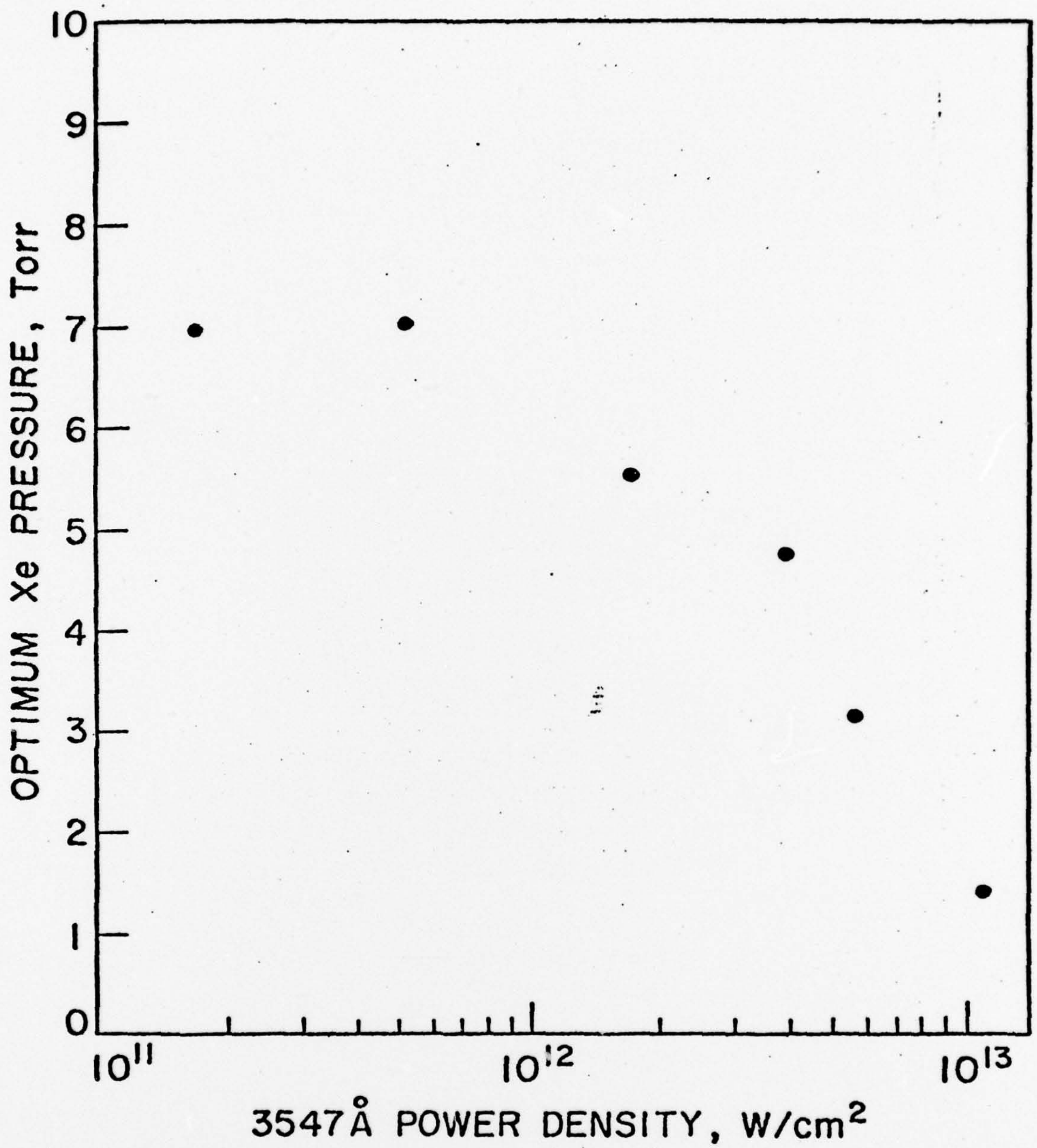

Fig. 2--Pressure of pure Xe required for peak conversion efficiency vs. fundamental power density.

From the measured efficiencies and Eq. (3) we find

$$\chi^{(3)}(-3\omega, \omega, \omega, \omega) = 2.3 \times 10^{-35} \quad \text{(esu)} \tag{5}$$

At higher P/A lower Xe pressures are required to optimize $\mathcal{E}$; at $P/A = 5.2 \times 10^{12}$ W/cm^2 , the optimum pressure has dropped by 2 to 3.5 torr, indicating that $\gamma = -6.3 \times 10^{-36}$ cm^5 W^{-1} or

$$\chi^{(3)}(-3\omega, 3\omega, -\omega, \omega) - \frac{1}{2}\chi^{(3)}(-\omega, \omega, -\omega, \omega) = 8.0 \times 10^{-35} \quad \text{(esu)} \tag{6}$$

For large incident power densities such that $|\gamma \frac{P}{A}| > |\alpha + \beta R|$ the Kerr-induced contribution will dominate the dispersion of the medium, and Eq. (3) shows that the conversion efficiency will be limited to a maximum value of

$$\mathcal{E}_{max} = 0.11 \left[\frac{\chi^{(3)}(3\omega, \omega, \omega, \omega)}{\chi^{(3)}(-3\omega, \omega, -\omega, \omega) - \frac{1}{2}\chi^{(3)}(-\omega, \omega, -\omega, \omega)} \right]^2 \tag{7}$$

For Xe our measurements indicate that $\mathcal{E}_{max} = 9 \times 10^{-3}$ independent of all experimental parameters; in particular, changing R only changes the power density at which the limit occurs. In addition, this maximum limiting efficiency can be obtained only by using an input pulse of uniform P/A in both space and time. Although this analysis has been done for the tight focusing case, Eq. (7) remains essentially unchanged for the plane wave case.

This analysis implies that the limiting efficiency of 2×10^{-4} in Fig. 1 is not a Kerr-induced limitation, but rather due to some other effect which becomes important at large P/A , such as multiphoton absorption or breakdown.

To circumvent this problem we shifted the Kerr dominated region to lower P/A by using an $R = 11$ Xe-Ar mixture; efficiencies up to 2×10^{-3} were observed, again limiting at about 5×10^{12} W/cm^2 .

The large magnitude of γ indicates that the mixed frequency term $\chi^{(3)}(-3\omega, 3\omega, -\omega, \omega)$ predominates in Eq. (2), since the calculated magnitude of $\chi^{(3)}(-\omega, \omega, -\omega, \omega)$ implies a value of γ about 10^3 below the observed value. The calculation of the mixed-frequency Kerr susceptibility in Xe involves two primary perturbation paths: $5p^6\ {}^1S \rightarrow 5d[3/2]^0_1 \rightarrow$ continuum $\rightarrow 5d[3/2]^0_1 \rightarrow 5p^6\ {}^1S$ and $5p^6\ {}^1S \rightarrow 5d[3/2]^0_1 \rightarrow 6p[5/2]_2 \rightarrow 5d[3/2]^0_1 \rightarrow 5p^6\ {}^1S$. It is difficult to determine the relative magnitude of the two contributions since neither the $5d[3/2]^0_1 \rightarrow$ continuum nor the $5d[3/2]^0_1 \rightarrow 6p[5/2]_2$ oscillator strengths have been measured. However, the negative sign of γ indicates that the path through the continuum provides the larger contribution.

Our results indicate that the ultimate 3547 Å $\rightarrow$ 1182 Å conversion efficiency in the Xe-Ar system is limited to less than 1% by a large mixed-frequency Kerr effect. It should be possible to overcome this limitation by adding an additional species with a compensating Kerr susceptibility; however, the species used must not interfere radically with the required net negative dispersion or the nonlinearity. Unfortunately, none of the inert gases seem to meet these requirements. One possible candidate appears to be Mg vapor; it is negatively dispersive and has a substantial nonlinearity for 3547 Å $\rightarrow$ 1182 Å generation.[6] We estimate that $\chi^{(3)}_{Mg}(-\omega, \omega, -\omega, \omega) \simeq -2 \times 10^{-33}$ esu ; thus a ratio of 25:1 of Xe:Mg should eliminate the Kerr-induced dispersion and still permit phasematching with Ar. We believe such three-component mixtures offer the possibility of a high power, high efficiency source of

1182 Å radiation. We note that the particular dispersion compensation scheme proposed here does not reduce the Kerr-induced beam focusing effects.[7]

We would like to acknowledge helpful discussions with S. E. Harris, and the technical assistance of Mr. Ben Yoshizumi.

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FIGURE CAPTIONS

1. Measured 3547 Å → 1182 Å peak conversion efficiency vs. fundamental power density in pure Xe.
2. Pressure of pure Xe required for peak conversion efficiency vs. fundamental power density.